Accurate Density and Viscosity Modeling of Non-Polar Fluids Based on the "*f-theory*" and a Non-Cubic EOS[1]

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ABSTRACT

Recently, the *f-theory*, a theory for viscosity modeling based on friction concepts of classical mechanics, has been introduced. This new theory allows accurate viscosity-pressure-temperature (η-p-T) modeling based on a van der Waals type of equation of state, one with a repulsive pressure term and an attractive pressure term. Thus, popular cubic equations of state (CEOS), such as the SRK and the PR, have been successfully applied in order to obtain accurate η-p-T models (even close to the critical region) of fluids such as n-alkanes, N_2, CO_2, between others, and some of their mixtures. However, even though it has been shown that a CEOS *f-theory* based model can accurately reproduce the viscosity behavior of, at least, non-polar fluids, the accuracy of the density predictions is still limited by the algebraic structure of the CEOS. In this work, a non-cubic van der Waals type of equation of state is introduced for the accurate modeling of both, the density and the viscosity behavior of selected non-polar fluids. The achieved accuracy, for both the density and viscosity fluid properties, is close to, or within, experimental uncertainty and applies to wide temperature and pressure ranges.

KEY WORDS: equation of state, f-theory, hydrocarbons, modeling, prediction, viscosity.

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1. INTRODUCTION

Recently, Quiñones-Cisneros et al. [1] proposed the friction theory (*f-theory*) for viscosity modeling and illustrated the capability of this theory by modeling the viscosity of n-alkanes from low to extremely high pressures and covering a reduced temperature range from 0.4 to 2.0. The *f-theory* is based on friction concepts of classical mechanics and the van der Waals theory of fluids. The main difference between the *f-theory* and other approaches to viscosity modeling is that the viscosity of dense fluids is considered as a mechanical rather than a transport property. In the *f-theory* the viscosity is linked to the pressure, which is the main mechanical variable, and, by use of a simple cubic equation of state, highly accurate viscosity estimations can be obtained from low to extremely high pressures. This is achieved regardless of the accuracy of the estimated density.

Due to their importance in the petrochemical industry, some of the better-investigated fluids in terms of viscosity and density are the n-alkanes. By smoothing experimental viscosity measurements, temperature and pressure tabulations of recommended viscosity values have been proposed by different researchers. One of the most relevant works in this area is the extensive compilation of recommended viscosity values for different pure fluids presented in 1979 by Stephan and Lucas [2]. However, based on new measurements, especially up to high pressures, Zéberg-Mikkelsen et al. [3] presented new tabulations of recommended viscosities of n-alkanes from methane to n-octadecane. The recommended viscosity values by Zéberg-Mikkelsen et al. [3] were obtained from experimental data using the *f-theory* for viscosity modeling. In addition, based on these tabulations, the constants of the friction parameters in the *f-theory* were also estimated for the Stryjek and Vera [4] modification of the Peng and Robinson cubic equation of state (EOS). Therefore, interpolations between the tabulated values can be performed with a good accuracy and the viscosity of n-alkanes mixtures can also be accurately predicted using the mixing rules for the friction parameters given in [1]. However, such a degree of accuracy is not extended to densities when a cubic EOS is used.

In order to be able to obtain accurate viscosity and density predictions, the *f-theory* has been extended to the recent modification of the Benedict-Webb-Rubin [5] EOS by Soave [6] (SBWR EOS). The SBWR EOS is capable of accurate density predictions for n-alkanes as large as n-C_{20} and in this work accurate viscosity modeling for almost the same number of n-alkanes has been achieved. However, since the SBWR is not a van der Waals type of EOS, i.e. it is not based on a repulsive and attractive pressure term, some considerations in that respect have been taken into account. In addition, a first attempt toward predicting the viscosity of n-alkanes mixtures is also considered in this work.

2. THE FRICTION THEORY

According to the *f-theory* [1], the total viscosity η of dense fluids can be separated into a dilute gas term η_0 and a friction term η_f,

$$\eta = \eta_0 + \eta_f \ . \tag{1}$$

The dilute gas term η_0 applies at the zero pressure limit of the gas phase and it can be accurately estimated with models such as the one proposed by Chung et al. [7]. This model is applicable for predicting the dilute gas viscosity of both polar and non-polar fluids over wide ranges of temperature with an absolute average deviation (AAD) of 1.5%. The dilute gas limit approximations are based on the Chapman-Enskog theory (Chapman et al. [8]) and the expression for the reduced collision integral obtained for the Lennard-Jones 12-6 potential by Neufeld et al. [9]. In micropoise (μP), the dilute gas model by Chung et al. is given by

$$\eta_0 = 40.785 \frac{\sqrt{MW\,T}}{v_c^{2/3}\Omega^*} F_c \quad , \tag{2}$$

where the following empirical equation is used to estimate the reduced collision integral:

$$\Omega^* = \frac{1.16145}{T^{*0.14874}} + \frac{0.52487}{\exp\left(0.77320T^*\right)} + \frac{2.16178}{\exp\left(2.43787T^*\right)}$$

$$- 6.435 \times 10^{-4} \, T^{*0.14874} \sin\left(18.0323T^{*-0.76830} - 7.27371\right) \tag{3}$$

with

$$T^* = \frac{1.2593T}{T_c} \quad . \tag{4}$$

In Eq. (2) the critical volume v_c is given in cm^3/mol, and for non-polar gases the F_c factor is empirically found to be

$$F_c = 1 - 0.2756\omega \quad . \tag{5}$$

In the case of the friction viscosity term η_f, Quiñones-Cisneros et al. [1] proposed an analogy between the Amontons-Coulomb friction law and the van der Waals repulsive and attractive pressure terms p_r and p_a. Thus, by means of three temperature dependent coefficients, η_f can be linked to the van der Waals repulsive and attractive pressure terms as follows:

$$\eta_f = \kappa_r \, p_r + \kappa_{rr} \, p_r^{\,2} + \kappa_a \, p_a \quad . \tag{6}$$

This approach, in conjunction with a simple cubic EOS, can provide highly accurate viscosity modeling of n-alkanes over wide ranges of temperature and pressure. Furthermore, by using simple mixing rules, the *f-theory* has also been found to give accurate mixture viscosity predictions without any need of viscosity binary parameters [1].

3. THE SBWR MODEL

Although the non-cubic EOS recently proposed by Soave [6] can provide accurate densities for n-alkanes with acentric factors as large as 0.9, the *f-theory* viscosity modeling based on this type of EOS is not straightforward. The *f-theory* is based on the van der Waals concept of a balance between the repulsive and the attractive pressure terms while a BWR type of EOS is not structured in this way. However, there are alternative ways by

which the *f-theory* can be adapted to an EOS that is not of the van der Waals type. For instance, if the total pressure in a given EOS is written as the addition of n terms,

$$p = \sum_{i=1}^{n} p_i \quad , \tag{7}$$

a straight forward extension of the *f-theory* is to write a model for the friction viscosity term as follows:

$$\eta_f = \sum_{i=1}^{n} \left(\kappa_{i,1} \, p_i + \kappa_{i,2} \, p_i^{\,2} \right) \quad . \tag{8}$$

In such a case, a term-by-term analysis may show that some of the second order terms may be neglected if they do not have an important contribution at high pressure. This approach has been tested for the SBWR EOS and extremely accurate modeling results have been obtained for pure components. However, if the viscosity of mixtures is predicted using the kind of simple mixing rules used with other *f-theory* models [1, 3, 10] the results are not as good and, in some cases, substantially large errors can be found. Therefore, if a model such as the one given in Eq. (8) is used for the prediction of mixture viscosities, adequate mixing rules have to be developed.

An alternative approach, to model the viscosity of pure fluids and to improve mixture viscosity predictions, is to group the terms of the non-van der Waals EOS into attractive-like and repulsive-like pressure terms in order to obtain a more consistent model with the *f-theory*. In the case of the SBWR EOS, this can be achieved by separating the equation into the following attractive-like and repulsive-like pressure terms:

$$p = p_a + p_r \quad , \tag{9}$$

where

$$p_a = B \, RT \, \rho^2 \tag{10}$$

and

$$p_r = \left(1 + D\rho^4 + E \, \rho^2 \left(1 + F \, \rho^2 \right) \exp\left(- F \, \rho^2 \right)\right) RT \, \rho \quad . \tag{11}$$

Figure 1 shows for n-heptane at a reduced temperature of 0.5 the p/p_c, p_a/p_c and p_r/p_c isotherms predicted by the SBWR EOS. Although it appears like equations (10) and (11)

have a correct qualitative attractive and repulsive performance, another major difference with the van der Waals type of EOS lies on the fact that the repulsive term is not a clearly dominating term at high pressure. This behavior is due to the lack of an excluded volume in the mathematical structure of an EOS such as the SBWR EOS. Thus, Eq. (6) cannot be directly applied since it was derived under the assumption that the repulsive term strongly dominates at high pressure. Therefore, if this assumption is removed, an extension of the *f-theory* to the SBWR EOS would give

$$\eta_f = \kappa_a\, p_a + \kappa_{aa}\, p_a^{\,2} + \kappa_r\, p_r + \kappa_{rr}\, p_r^{\,2} \quad , \tag{12}$$

where κ_a, κ_r, κ_{aa} and κ_{rr} are the corresponding temperature dependent linear and quadratic friction parameters.
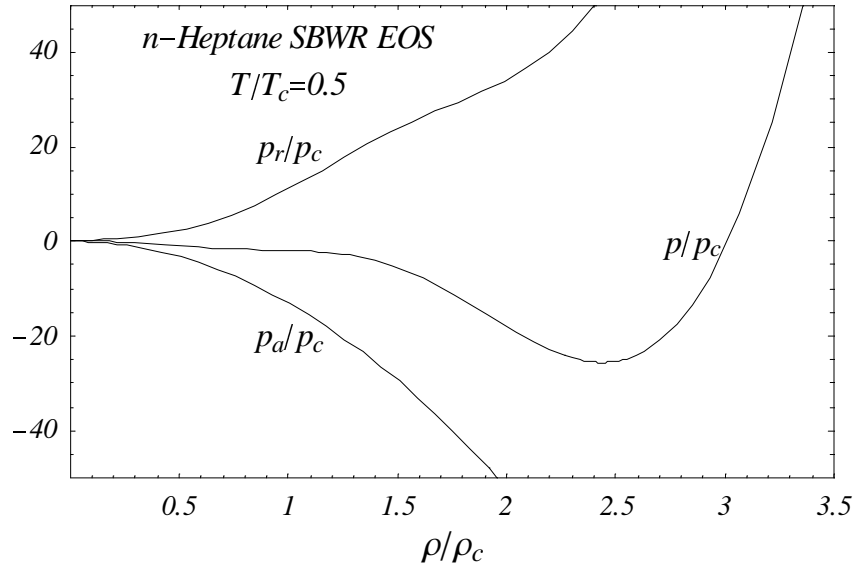
Figure 1. The SBWR n-heptane reduced pressure, reduced attractive pressure term and reduced repulsive pressure term as a function of the reduced density at a reduced temperature of 0.5.

For the temperature dependency of the friction parameters it has been found that a regular second order polynomial on the inverse of the reduced temperature gives accurate results. On the other hand, similarly to the quadratic term of the van der Waals *f-theory* models [1], an exponential term, which vanishes as temperature increases, is required for an accurate temperature dependency description of the second order terms. This results in the following empirical models for the temperature dependency of the friction terms:

$$\kappa_r = a_0 + a_1 T_r^{-1} + a_2 T_r^{-2} \quad , \tag{13}$$

$$\kappa_a = b_0 + b_1 T_r^{-1} + b_2 T_r^{-2} \tag{14}$$

$$\kappa_{rr} = c_2 \left(\exp\left[2T_r^{-1}\right] - 1 \right) \tag{15}$$

and

$$\kappa_{aa} = d_2 \left(\exp\left[2T_r^{-1}\right] - 1 \right) \quad , \tag{16}$$

where

$$T_r = \frac{T}{T_c} \quad . \tag{17}$$

4. VISCOSITY MODELING OF PURE NORMAL ALKANES

In order to derive general models for n-alkanes, an overall least squares (LS) fitting of the recommended viscosities reported by Zéberg-Mikkelsen et al. [3] has been performed for the *f-theory* SBWR model. In addition, a LS fitting of the n-nonane and n-undecane Stephan and Lucas [2] recommended data has also been carried out. The SBWR EOS has been used as described by Soave with respect to all of the parameters and parametric laws presented in [6]. The critical temperature, critical pressure and acentric factor values necessary in the SBWR EOS have been taken from the table of recommended constants by Stryjek and Vera [4] and the molecular weight values from Reid et al. [11]. For consistency, the critical volumes required in Eq. (2) have been estimated with the empirical equation for the critical compressibility used by Soave in the SBWR EOS [6, 12],

$$Z_c = 0.2908 - 0.099\,\omega + 0.04\,\omega^2 \quad . \tag{18}$$

Table I. Parameters for the *f-theory* with the SBWR EOS.

	a_0 (μP/bar)	a_1 (μP/bar)	a_2 (μP/bar)	b_0 (μP/bar)	b_1 (μP/bar)	b_2 (μP/bar)	c_2 (μP/bar^2)	d_2 (μP/bar^2)
Methane	0.123927	0.112587	-0.0185216	-0.489896	0.539714	-0.251465	5.63508×10^{-6}	-5.66315×10^{-6}
Ethane	0.163212	0.12666	0.126043	-0.433616	0.308836	-0.00275856	8.90028×10^{-6}	-7.82672×10^{-6}
Propane	1.03707	-1.43503	0.914526	0.363672	-1.27786	0.749809	1.33420×10^{-6}	-1.12798×10^{-6}
n-Butane	0.547583	-0.0753582	0.257617	0.0244887	-0.154573	0.137811	2.74777×10^{-6}	-2.32236×10^{-6}
n-Pentane	1.54698	-1.30029	0.504511	2.20018	-3.92745	1.67632	5.75590×10^{-6}	3.69405×10^{-6}
n-Hexane	0.238079	0.863081	-0.353355	-0.522078	0.655814	-0.35482	9.54554×10^{-6}	-8.07294×10^{-6}
n-Heptane	0.713028	0.362262	-0.186094	-0.250222	0.605241	-0.453361	1.14152×10^{-5}	-1.10397×10^{-5}
n-Octane	-1.07202	4.0199	-1.95067	-0.483369	1.69208	-1.18746	1.62559×10^{-5}	-1.13425×10^{-5}
n-Nonane	8.18267	-12.2254	5.37905	7.14981	-12.6186	5.50978	2.71259×10^{-6}	1.92787×10^{-6}
n-Decane	-0.100514	2.54595	-1.28388	-0.794063	1.87837	-1.17878	2.03760×10^{-5}	-1.63155×10^{-5}
n-Undecane	9.9027	-12.6375	4.8085	7.90016	-11.1857	4.03575	1.26641×10^{-5}	-1.09130×10^{-5}
n-Dodecane	5.87858	5.41329	-5.77504	10.8100	-1.92149	-3.70594	3.71540×10^{-5}	-2.99154×10^{-5}
n-Tridecane	22.7060	-16.8488	2.50591	29.5034	-24.5543	4.10497	2.80472×10^{-5}	-2.42730×10^{-5}
n-Tetradecane	98.1950	-82.0220	14.4688	112.839	-100.306	19.6573	4.01647×10^{-5}	-2.94785×10^{-5}
n-Pentadecane	9.89108	8.48017	-9.28628	16.9267	-1.372	-6.54706	5.16062×10^{-5}	-4.27417×10^{-5}
n-Hexadecane	-47.4218	87.1473	-36.6045	-29.5602	65.7989	-30.8006	7.50760×10^{-5}	-6.38778×10^{-5}
n-Octadecane	268.725	-260.773	61.415	348.145	-352.000	87.5166	4.51233×10^{-5}	-8.31360×10^{-6}

Table II. Overall performance of the *f-theory* SBWR model for pure alkanes.

|	T/T_c Range	p/p_c Range	AAD (%)	Point of Max. Dev. T/T_c	p/p_c	Max. Dev. (%)
Methane	0.55-2.50	0-20.0	0.95	1.0	1.05	6.44
Ethane	0.35-1.60	0-11.3	2.01	1.0	0.60	11.4
Propane	0.30-1.29	0-13.0	2.72	1.1	0.8	10.3
n-Butane	0.35-1.04	0-18.0	1.74	1.0	0.6	16.1
n-Pentane	0.64-1.16	0-30.0	1.14	1.1	0.8	6.78
n-Hexane	0.54-1.08	0-35.0	1.38	1.0	0.95	12.4
n-Heptane	0.56-1.00	0-35.0	0.67	1.0	0.95	8.65
n-Octane	0.50-1.00	0-40.0	1.09	1.0	0.95	7.90
n-Nonane	0.50-0.79	0-30.2	0.66	0.64	4.4	3.71
n-Decane	0.45-0.76	0-48.0	0.49	0.65	8.0	1.80
n-Undecane	0.47-0.81	0-25.4	0.45	0.81	20.3	2.98
n-Dodecane	0.45-0.60	0-55.0	0.52	0.6	54.8	1.62
n-Tridecane	0.44-0.53	0-60.0	0.35	0.525	60.0	1.25
n-Tetradecane	0.42-0.54	0-60.0	0.63	0.51	0.2	2.09
n-Pentadecane	0.44-0.58	0-65.0	0.65	0.575	65.0	2.15
n-Hexadecane	0.41-0.52	0-70.0	1.24	0.46	70.0	3.53
n-Octadecane	0.42-0.55	0-80.0	0.94	0.51	0.2	3.19

The *f-theory* SBWR friction constants, required in equations (13) through (16), have been obtained through a LS fitting of the considered n-alkanes data, as described above, and they are given in Table I. In Table II the temperature and pressure ranges of the modeled data together with the absolute average deviation (AAD), the point of maximum deviation and the maximum deviation value are given. The results reported in Table II show a very accurate model performance in all cases. In addition, all of the maximum deviations are also within experimental uncertainty. This follows from the fact that for all light n-alkanes, from methane to n-octane, the point of maximum deviation is close to the critical point where the viscosity derivative with respect to the pressure diverges. For all of the remaining dense hydrocarbons, the maximum deviation value is not larger than the experimental uncertainty of the original data.

Figures 2, 3 and 4 show th *f-theory* SBWR model results for a light alkane (propane), an intermediate one (n-heptane), and a dense one (n-pentadecane). The propane results (Figure 2) show a good agreement with the data. In addition, the propane predictions show

a good, consistent and stable performance, even for the low temperature extrapolated pressure cases where calculations have been made up to 600 bar. The same performance remarks also apply for the n-heptane results depicted in Figure 3, where the maximum pressure reaches 1600 bar. Finally, the n-pentadecane results depicted in Figure 4 also show a good model performance within the entire data temperature and pressure ranges.
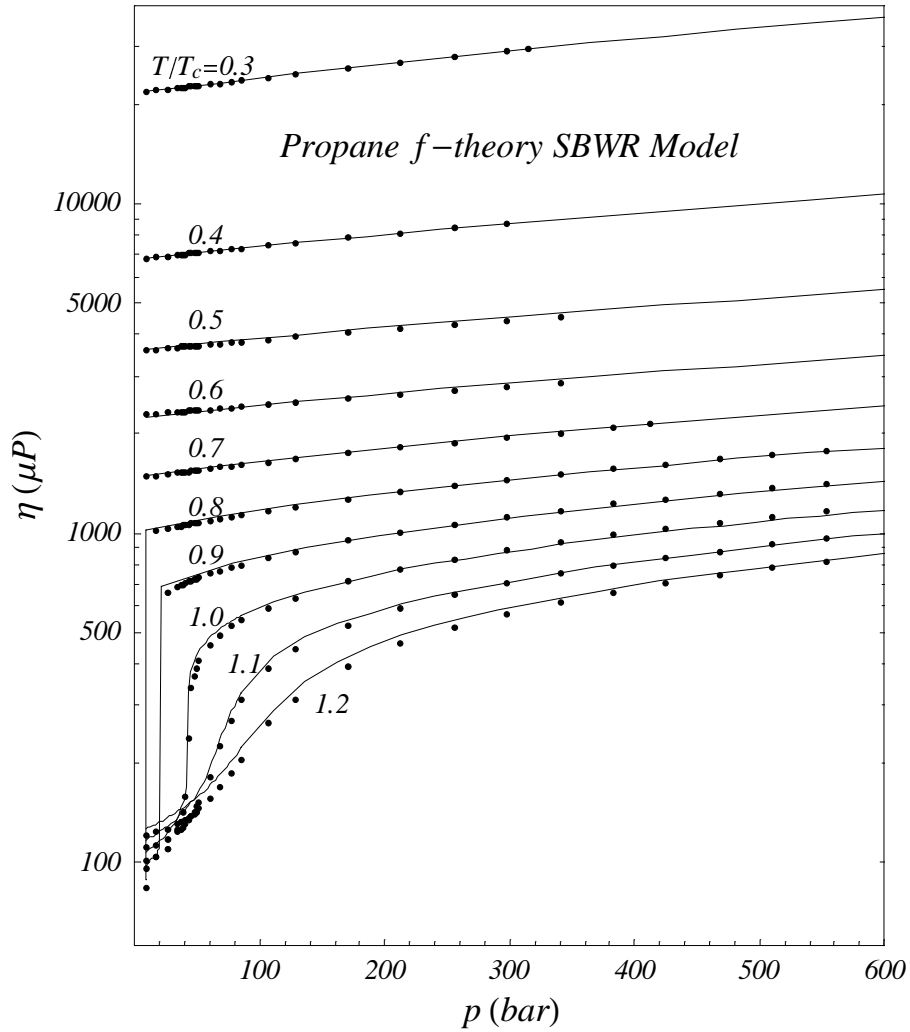
Figure 2. Propane viscosity results for the *f-theory* SBWR model (solid curves) along with the recommended data by Zéberg-Mikkelsen et al. [3] (points).
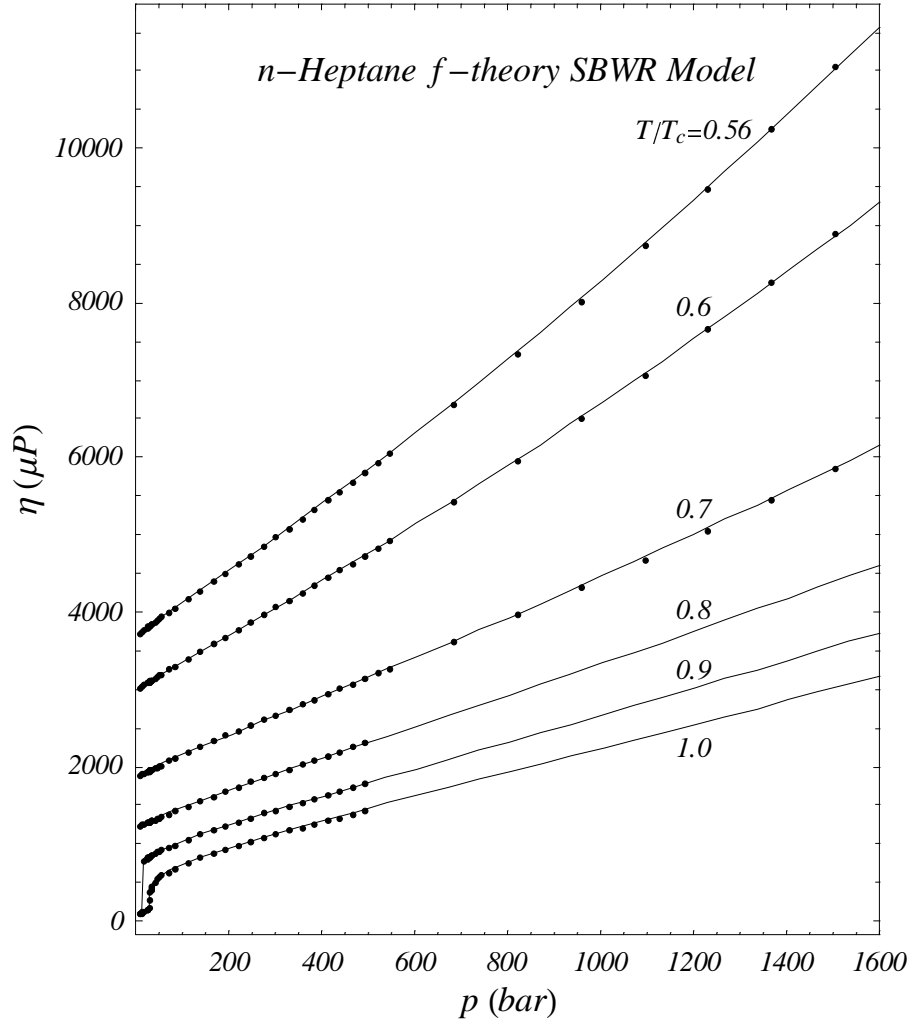
Figure 3. n-Heptane viscosity results for the *f-theory* SBWR model (solid curves) along with the recommended data by Zéberg-Mikkelsen et al. [3] (points).
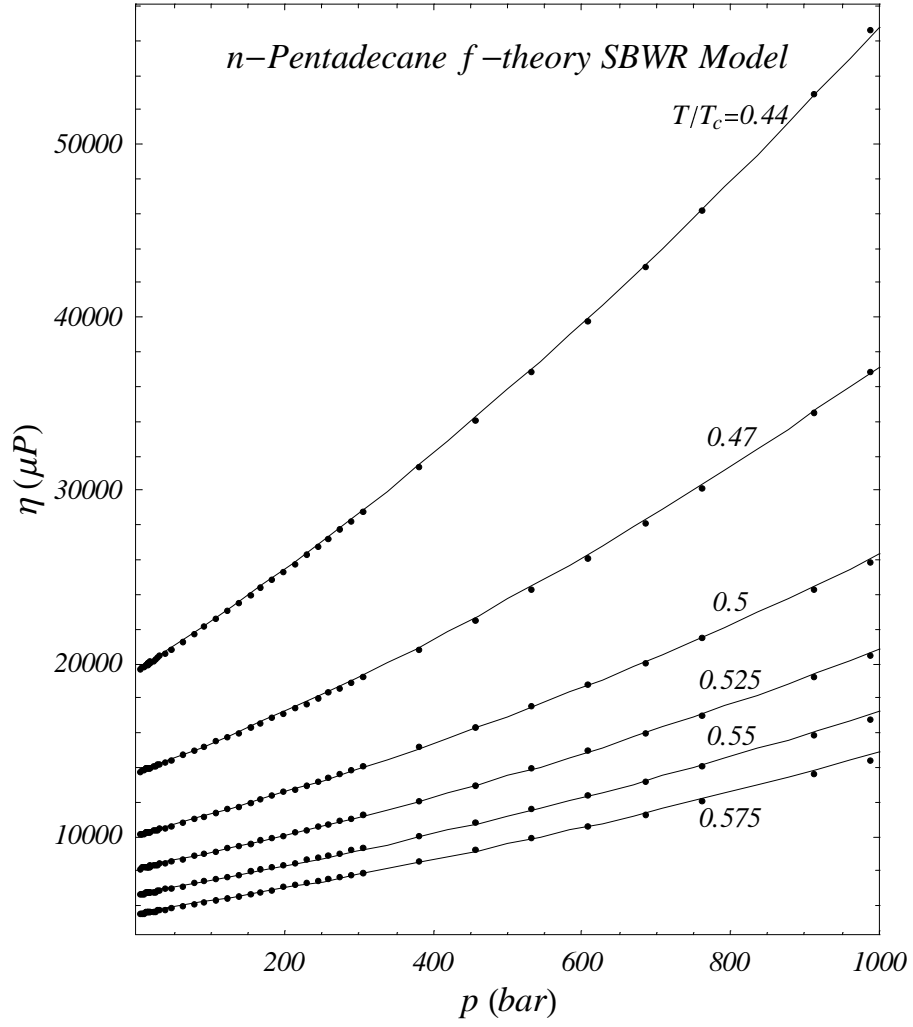
Figure 4. n-Pentadecane viscosity results for the *f-theory* SBWR model (solid curves) along with the recommended data by Zéberg-Mikkelsen et al. [3] (points).

5. VISCOSITY MODELING OF NORMAL ALKANES MIXTURES

In the *f*-theory original article [1] it was shown that some simple mixing rules for the friction parameters provide good results for the prediction of mixture viscosities. In this work it has been found that, for the *f-theory* SBWR, a good performance is also obtained with similar mixing rules. Thus, in the case of mixtures, the viscosity is given by

$$\eta_{mx} = \eta_{0,mx} + \eta_{f,mx} \quad , \tag{19}$$

where the subscript *mx* indicates the corresponding mixture property. Here, the mixture dilute gas limit is calculated by

$$\eta_{0,mx} = \exp\left[\sum_{i=1}^{n} x_i \ln(\eta_{0,i}) \right] \quad . \tag{20}$$

In all cases, the subscript *i* refers to the corresponding pure component of an *n* components mixture. For the *f-theory* SBWR model, the mixture friction contribution term is given by

$$\eta_{f,mx} = \kappa_{r,mx}\, p_r + \kappa_{a,mx}\, p_a + \kappa_{rr,mx}\, p_r^2 + \kappa_{aa,mx}\, p_a^2 \quad , \tag{21}$$

where $\kappa_{r,mx}$, $\kappa_{a,mx}$, $\kappa_{rr,mx}$ and $\kappa_{aa,mx}$ are the viscous friction parameters for the mixture. Hence, for the viscous friction parameters the following simple mixing rules have been found to deliver satisfactory results:

$$\kappa_{r,mx} = \sum_{i=1}^{n} x_i\, \kappa_{r,i} \quad , \tag{22}$$

$$\kappa_{a,mx} = \sum_{i=1}^{n} x_i\, \kappa_{a,i} \quad , \tag{23}$$

$$\kappa_{rr,mx} = \sum_{i=1}^{n} x_i\, \kappa_{rr,i} \tag{24}$$

and

$$\kappa_{aa,mx} = \sum_{i=1}^{n} x_i\, \kappa_{aa,i} \quad . \tag{25}$$

Table III shows the AAD results for the mixture viscosity predictions of different kind of n-alkanes mixtures. In general, it can be appreciated that the accuracy of the mixture predictions is satisfactory. A larger deviation is found for the methane+n-decane system, which may be due to a combination of factors that include the uncertainty in the experimental data, the large acentric difference between the methane and the n-decane, and the fact that, as suggested by Soave [6], all binary interaction parameters have been set to zero. Figure 5 illustrates the prediction results for the quaternary $n-C_{10}+n-C_{12}+n-C_{14}+n-C_{16}$ mixture.

Table III. Overall performance for viscosity predictions of hydrocarbon mixtures along with the data source reference.

	Reference	AAD (%)	Max. Dev. (%)
$C_1 + C_2$	[13]	3.91	14.56
$C_1 + C_3$	[14]	4.21	11.30
$C_1 + n-C_4$	[15]	2.42	9.78
$C_1 + n-C_6$	[16]	4.06	9.70
$C_1 + n-C_{10}$	[17]	15.75	19.88
$n-C_5+n-C_8$	[18]	4.56	11.91
$n-C_5+n-C_{10}$	[19]	4.84	17.79
$n-C_6 + n-C_7$	[20]	0.91	1.52
$n-C_7+n-C_8$	[21]	2.23	7.56
$n-C_7+n-C_9$	[20]	0.81	2.62
$n-C_8+n-C_{10}$	[22]	3.46	6.95
$n-C_{10}+n-C_{16}$	[23]	4.73	9.58
$n-C_5+n-C_8+n-C_{10}$	[24]	2.41	11.28
$n-C_{10}+n-C_{12}+n-C_{14}+n-C_{16}$	[23]	5.68	9.86
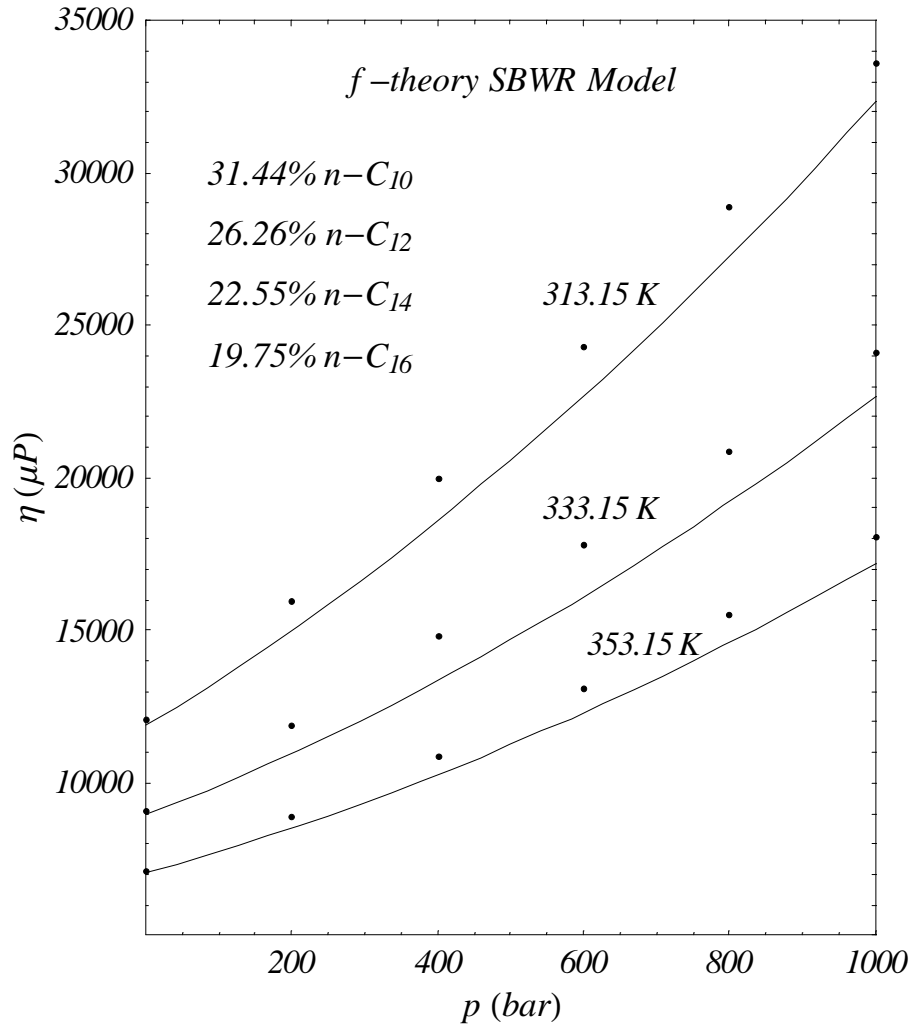
Figure 5. Mixture viscosity predictions with the *f-theory* SBWR model (solid curves) for the n-C_{10}+n-C_{12}+n-C_{14}+n-C_{16} quaternary system along with the data by Ducoulombier et al. [23] (points).

6. CONCLUSIONS

In this work the *f-theory* has been extended to the SBWR EOS, a Benedict-Webb-Rubin type of EOS recently proposed by Soave [6]. Thus, highly accurate viscosity modeling of n-alkanes up to n-octadecane has been achieved. In addition, the viscosity of several n-alkane mixtures has also been predicted with a good degree of accuracy. In the case of pure n-alkanes, the viscosity modeling results obtained in this work are as accurate as the results previously obtained with cubic EOS. Therefore, an advantage of this model is the simultaneous achievement of accurate viscosity and density modeling.

In general terms, this work illustrates how an equation of state, different to a van der Waals type of EOS, can be used to generate an accurate viscosity model. Clearly, there are many other EOS that are used in different industries, a large amount of them are cubic EOS but many others are not. Therefore, following the lines presented in this and previous related works [1, 3, 10], it may be possible to derive *f-theory* viscosity models based on different available equations of state in order to extend their capabilities to viscosity modeling.

Finally, although the general accuracy of the mixture viscosity predictions is satisfactory, the *f-theory* SBWR mixture results appear to be less accurate than the results obtained with *f-theory* models based on cubic EOS [1, 3, 10]. This may be due to the better repulsive and attractive structure of the cubic EOS compared to a BWR type of EOS. Nonetheless, it may also be due to the fact that in the SBWR EOS no binary interaction parameters were used. This appears to be a good choice for most of the studied mixtures however, the mixtures that have presented the largest AAD's were those compose of molecules with a large acentric difference such as methane+n-decane. Therefore, the use of binary parameters in the SBWR EOS may result in better viscosity predictions. In fact, the most acentric mixture considered by Soave [6] was methane+pentane and to that acentric range the viscosity predictions of the *f-theory* SBWR model are rather

accurate. Furthermore, if a binary parameter is used for the methane+n-decane mixture, the AAD reduces to 8.56% for a binary parameter of –0.1 and it reaches a 5.37% AAD minimum when the binary parameter reaches a value of around –0.2. Thus, if the SBWR EOS is to be used for phase and viscosity modeling of mixtures with large acentric differences, binary parameters may also be required.

ACKNOWLEDGMENTS

LIST OF SYMBOLS

Latin Letters

T	=	absolute temperature
T_c	=	critical temperature
p	=	total pressure
p_c	=	critical pressure
p_a	=	van der Waals attractive pressure term
p_r	=	van der Waals repulsive pressure term
v_c	=	critical volume
x_i	=	mole fraction of component i

Greek Letters

η	=	total viscosity
η_0	=	dilute gas viscosity
η_f	=	friction viscosity
κ_a	=	linear attractive viscous friction coefficient
κ_{aa}	=	quadratic attractive viscous friction coefficient
κ_r	=	linear repulsive viscous friction coefficient
κ_{rr}	=	quadratic repulsive viscous friction coefficient
ω	=	Pitzer's acentric factor

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